United States Patent Office

3,829,354

Patented Aug. 13, 1974

3,829,354

HYDROGEN SULFIDE-MODIFIED EPOXY RESINS AND FLEXIBLE LAMINATES THEREFROM

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No Drawing. Continuation-in-part of abandoned application Ser. No. 185,893, Oct. 1, 1971. This application Feb. 23, 1973, Ser. No. 335,379

Int. Cl. C08g *45/06;* B32b *27/38;* B32b *27/04*

U.S. Cl. 161—88 6 Claims

ABSTRACT OF THE DISCLOSURE

Mixtures of the diglycidyl ethers of a bisphenol such as bisphenol A and a diglycidyl ether of an aliphatic polyhydroxyl containing compound such as the diglycidyl ether of neopentyl glycol are modified with hydrogen sulfide to produce epoxy resins which when cured with an aminated polyglycol are useful in the preparation of flexible laminates.

This application is a continuation-in-part of our copending application Ser. No. 185,893, filed Oct. 1, 1971, now abandoned.

This invention relates to new epoxy resin compositions, cured products and flexible laminates prepared therefrom.

Background of the Invention

The flexible electrical laminate industry is a relatively new art. Until a few years ago, the 1/16 inch hardboard had been the standard product in the electrical laminate field. In the last two to five years, a tremendous technicological boom has occurred with the advent of the ultrathin and multilayer board. The advantages of flexible circuitry include great savings in space, weight, and cost of assembly.

Relatively few materials are available which are suitable for use in flexible laminates. In general, three material types have been used for most flexible circuit device fabrication; FEP (polymers of fluorinated ethylene such as Teflon or fluorinated propylene copolymers thereof, polyester film (Mylar), and polyimide (Kapton) film. These are all unreinforced films. There is a need for reinforced films also. The industry is looking for a flexible epoxy resin/hardner system to be impregnated in a suitable substrate such as glass cloth, Reemay (spun Mylar), Nomex nylon paper, etc. In the case of FEP substrates, copper is heat bonded directly to the film without cladding adhesives. However, the thermal stability is bad with FEP. The 520° F. soldering temperature approaches the melting point of FEP. During soldering, complete loss of copper adhesion can occur as well as physical distortion of the circuitry device. Other FEP disadvantages include cold flows and low tensile strength. FEP advantages include excellent electrical properties, flame retardancy and good peel strength.

Polyester film substrates are metal cladded using a cladding adhesive. Excessive heat during soldering (520° F.) can cause film disorientation with a resultant loss of desirable properties. Mylar degrades at 425° F. When polyester film is dipped in molten solder, it immediately melts. Another disadvantage besides the poor thermal stability is the minimal copper peel strength. Advantages include toughness, good electrical properties, good solvent and water resistance, and good flexibility.

At the present, no one is able to pass the rigorous thermal stability requirement except through the use of high priced polyimide (Kapton). Kapton film costs around $25.00 per pound.

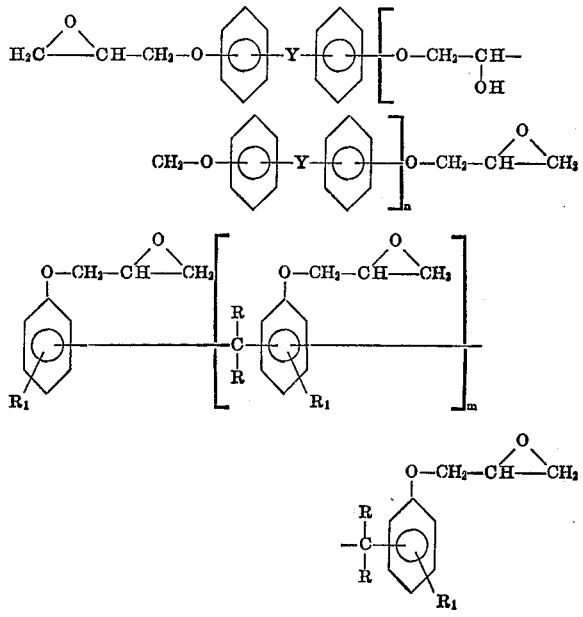
The compositions of the present invention when cured with a polyglycol diamine give very good flexible laminate properties. Reemay (spun Mylar), glass, or Nomex nylon paper can be impregnated with the above formulation to give good properties.

Epoxy resins have been modified previously by reaction with hydrogen sulfide as taught in U.S. 2,633,458; however the epoxy resin compositions of the present invention have not previously been prepared. It is these hydrogen sulfide modified epoxy resin compositions of the present invention that make the flexible epoxy resin electrical laminates of the present invention possible. The examples herein will demonstrate that laminates prepared from the hydrogen sulfide modified epoxy resins of the prior art possess certain deficiencies such as one or more of the following, not readily capable of being B-staged, poor flexibility, delamination, and failure of a crease test hereinafter described.

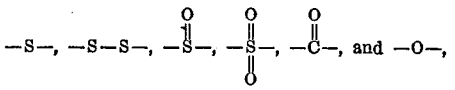
Detailed Description of the Invention

The epoxy resin compositions of the present invention comprise the reaction product of hydrogen sulfide with an epoxy resin mixture comprising

(A) from about 40 to about 85 percent and preferably from about 50 to about 75 percent by weight based upon the combined weights of A and B of an epoxy resin represented by the general formulae



wherein each Y is independently a divalent hydrocarbon radical having from about 1 to about 6 carbon atoms,



each R is independently hydrogen, methyl or ethyl, each R_1 is independently hydrogen or an alkyl group having from 1 to about 4 carbon atoms, m has an average value of from about 0.01 to about 2, and n has an average value of from about 0 to about 0.2.

(B) from about 15 to about 60 percent and preferably from about 25 to about 50 percent by weight based upon the combined weights of A and B of the diglycidyl ether of an aliphatic polyhydroxyl containing compound